Fermi surface shrinking and interband coupling in iron-based pnictides

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Recent measurements of Fermi surface with de Haas-van Alphen oscillations in LaFePO showed a shrinking of the Fermi pockets with respect to first-principle calculations, suggesting an energy shift of the hole and electrons bands with respect to local density approximation. We show that these shifts are a natural consequence of the strong particle-hole asymmetry of electronic bands in pnictides, and that they provide an indirect experimental evidence of a dominant interband scattering in these systems.

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A new challenge in the field of condensed matter is represented by the recent discovery of high-T_c superconductivity in the iron-based pnictide family [1]. First-principle calculations have soon identified four main bands at the Fermi level with two-dimensional character: two hole-like pockets around the Γ point and two electron-like pockets around the M point [2, 3, 4, 5, 6, 7]. A fifth three-dimensional band crossing the Fermi level at the Γ point is sometimes predicted by Local Density (LDA) calculations [2, 3], even if its position with respect to the chemical potential is strongly affected by the interlayer distance. Since determining the Fermi surface topology is the first step toward the understanding of these new materials, a lot of experimental work has been devoted to its investigation. A momentum-resolved mapping of the dispersion of the occupied quasi-particle states in the normal state and in the superconducting one has been provided by angle-resolved photoemission spectroscopy (ARPES), both in 1111 [8] and 122 systems [9, 10, 11, 12, 13]. An alternative technique probing the Fermi surface topology, which is not momentum resolved but has the advantage of being a bulk probe, is based on de Haas-van Alphen (dHvA) magnetization measurements, which allow one to estimate the size of the Fermi areas and the effective mass m^* for each Fermi sheet [14, 15, 16, 17, 18]. Although an overall qualitative agreement is found between LDA calculations and these experiments, interesting discrepancies still remain. In general, we can distinguish between *high energy* and *low energy* discrepancies. ARPES data in $Ba_{0.6}K_{0.4}Fe_2AS_2$ [9, 10] and LaFePO [8] for instance find overall bandwidths a factor 2 smaller than the LDA calculations, possibly related to the static electronic correlation [19]. On the other hands, the ARPES observation of Fermi velocities a factor 4 larger than the LDA calculated low-energy ones [10] points out a relevant role of dynamical quantum renormalization effects associated with a low-energy boson-mediated interaction.

Besides the bandwidth renormalization, a second striking result emerging from dHvA is a substantial shift of the bands with respect to the Fermi level when compared to LDA calculations. Indeed, the accurate determination of the Fermi surface areas provided by dHvA gives values smaller then those expected by LDA. Such discrepancy can be accounted for assuming a shift of the LDA bands. Notably, such shifts have different signs in hole and electron bands, being downward for the hole-band and *upward* for the electron ones[14, 15]. This effect seems to persist to higher energies, as one can infer from ARPES, which gives a systematical reduction of the energy difference between the bottom of the electron bands and the top of the hole bands with respect to the values predicted by LDA [10, 11, 12, 13]. Even though a certain degree of inaccuracy could be present in density-functional theory calculations, the persistent observations of such shifts suggests a robust feature in these materials whose origin is at the moment unknown.

In this Letter we present a comprehensive explanation for the origin of the band shifts observed in dHvA experiments. We show that they are a natural consequence of the multiband character of these systems and of the strong particle-hole asymmetry of the bands, which induce finite-band self-energy effects that are usually irrelevant for the half-filled single-band case. We also show that a simple analysis of the dHvA data provides evidence that the dominant interaction in these systems is the interband one between hole and electronic states, as it has been already argued on the basis of the nesting between hole and electron Fermi sheets [7, 20, 21, 22, 23]. More precisely, our calculations give an estimate of the interband coupling V of the order $V \approx 0.46$ eV, which gives rise also to a mass enhancement $Z_\alpha \approx 1.4$ for each band.

We start our analysis by discussing first a generic multiband system, where the electrons are coupled to a bosonic mode with local propagator $D(\omega_l) = \int d\Omega 2\Omega B(\Omega)/(\Omega^2+\omega_l^2)$, and where $B(\Omega)$ is the density of states of the bosonic excitations. The self-energy in the Matsubara space can be written as:

$$\Sigma_\alpha(i\omega_n) = -T\sum_{m,\beta} V_{\alpha,\beta} D(\omega_n-\omega_m) G_\beta(i\omega_m), \quad (1)$$

where α, β are band indexes, $V_{\alpha,\beta}$ ($V_{\alpha,\beta} = V_{\beta,\alpha}$) is the multiband interaction, which for a retarded interaction is always positive ($V_{\alpha,\beta} > 0$), and $G_\alpha(z)$ is the local Green's function for the α band, namely:

$$G_\alpha(z) = \int_{E_{\min,\alpha}}^{E_{\max,\alpha}} d\epsilon N_\alpha(\epsilon) \frac{1}{z - \epsilon - \Sigma_\alpha(z) + \mu}. \quad (2)$$

Here $N_\alpha(\epsilon)$ is the electronic density of states of the α band, with upper and lower band edges $E_{\max,\alpha}$ and $E_{\min,\alpha}$, respectively, and we drop the spin index since it does not play any role in the following.

In the conventional Eliashberg analysis [24], one usually assumes that the distance of the chemical potential from the band edges is much larger than the typical boson energy scale, so that one can approximate the DOS with its value at the Fermi level, $N_\alpha(\epsilon) \approx N_\alpha(\mu)$, and one can extend the integration limits over ϵ in Eq. (2) to $\pm\infty$. In this way one is enforcing the particle-hole symmetry and the Matsubara self-energy $\Sigma_\alpha(i\omega_n)$ is purely imaginary. Enforcing implicitly the particle-hole symmetry can be however quite dangerous in systems as the iron-pnictides where each band is strongly away from the half-filling. As a byproduct of taking into account the particle-hole *asymmetry*, the self-energy acquires a finite real part $\chi_\alpha(i\omega_n) \equiv \mathrm{Re}\Sigma_\alpha(i\omega_n) \neq 0$, whose low energy limit $\chi_\alpha = \chi_\alpha(i\omega_{n=0})$ gives rise to a band shift that can be in general different in each band. While this effect is usually disregarded in single-band systems, because it can be absorbed in a redefinition of the chemical potential, in the multiband case it can lead to observable relative shifts of the various bands with respect to the Fermi level.

Before undertaking a fully numerical solution of Eqs. (1)-(2), it is instructive to consider the result of the lowest order perturbation theory, where the Green's function $G_\beta(i\omega_m)$ in Eq. (1) is taken to be the non-interacting one with $\Sigma_\beta(i\omega_m) = 0$. We shall assume also purely two-dimensional parabolic bands, so that $N_\alpha = 1/(E_{\max,\alpha} - E_{\min,\alpha})$ and we consider for the moment, for simplicity, a Einstein mode $B(\Omega) = (\omega_0/2)\delta(\Omega - \omega_0)$. We get thus the analytical expression (for $T \approx 0$):

$$\chi_\alpha = -\frac{\omega_0}{2} \sum_\beta V_{\alpha,\beta} N_\beta \ln \left| \frac{\omega_0 - \mu + E_{\max,\beta}}{\omega_0 + \mu - E_{\min,\beta}} \right|. \quad (3)$$

In most cases, the exchanged boson energy is the lowest energy scale in the system, so that

$$\chi_\alpha \approx -\frac{\omega_0}{2} \sum_\beta V_{\alpha,\beta} N_\beta \ln \left| \frac{E_{\max,\beta} - \mu}{E_{\min,\beta} - \mu} \right|. \quad (4)$$

Eq. (4) gives correctly $\chi_\alpha = 0$ in the case of particle-hole symmetry $|E_{\max,\beta} - \mu| = |E_{\min,\beta} - \mu|$. On the other hand, in an electron-like band $|E_{\max,\beta} - \mu| > |E_{\min,\beta} - \mu|$, while the opposite inequality applies for a hole-like band. Let us consider now a single band system. In this case the band shift $\chi_{\alpha,0}$ is compelled to be positive (upward) for a hole-system and negative (downward) for an electron-like band. Such observation seems at odds with what reported in LaFePO.

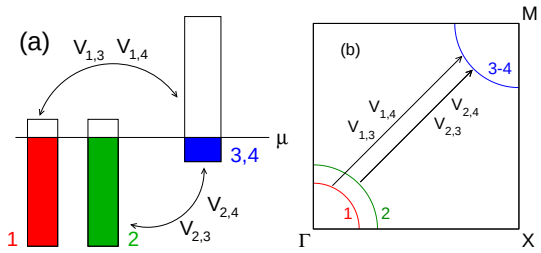

FIG. 1: (Color online). Sketch of our band structure model (a) and of the corresponding Fermi surfaces (b). We also show the only non-zero interband couplings.

Things are however more subtle in multiband systems, where the shift of each band depends also on the contribution of *all the other bands* weighted with the corresponding interband coupling. This effect is particularly important in pnictides where the dominant coupling is thought to be the spin-mediated interband interaction. In such a situation, the particle-hole asymmetry of the electron bands is responsible for the downward shift of the hole-like bands which, vice versa, give rise to the upward shift of the electron ones. We would like to stress that these argumentations do not depend on the band or interaction details. In full generality *the simple observation of a upward shift of the electron bands and the downward shift of the hole-like ones is a direct experimental evidence in these compounds of the dominance of the interband coupling on the intraband one, which would produce the opposite scenario.*

Within this framework, a more detailed analysis of the dHvA measurements in LaFePO [14] can provide an useful insight into the *strength* of the interband coupling in pnictides. Following Ref. [25] we consider four bands ($1, 2$ hole-like and $3, 4$ electron-like) and we assume, because of the nesting properties, a purely interband scattering connecting hole and electron bands, namely $V_{1,3}, V_{1,4}, V_{2,3}, V_{3,4}$, (see Fig. 1). Note that, unlike to other pnictides, all the Fermi areas in LaFePO are quite comparable, so that we can expect that interband scattering $V_{\alpha,\beta}$ does not depend much on the band index, and we can assume, as a first approximation, $V_{1,3} = V_{1,4} = V_{2,3} = V_{3,4} = V$. We model each band with a purely two-dimensional parabolic dispersion, and we assume for simplicity bands $3, 4$ to be completely degenerate. The band parameters are extracted from the LDA calculations [2]. More explicitly, focusing on the $k_z = 0$ plane, we determine the Fermi vectors $k_{\mathrm{F},\alpha}$ from the Γ-X cut for the hole-bands, and from the M-X cut for the electron ones. Setting for convention the Fermi level $\mu = 0$, we also take from LDA calculations the nearest

band index	m_α/m_e	$E_{\max,\alpha}$ (eV)	$E_{\min,\alpha}$ (eV)	N_α (eV^{-1})	χ_α (meV)	m^*_α/m_e
LDA calculations LaFePO						
1	0.58	0.205	-5.031	0.191	-53	1.0
2	1.14	0.205	-2.462	0.375	-53	2.0
3,4	0.79	3.551	-0.295	0.260	62	1.4
Renormalized LDA calculations LaFePO						
1	1.16	0.102	-2.516	0.382	-26.5	1.6
2	2.28	0.102	-1.231	0.750	-26.5	3.2
3,4	1.58	1.776	-0.147	0.520	31	2.3

TABLE I: Microscopic band parameters extracted from unrenormalized LDA calculations [2] (top), and renormalized ones (bottom). Also shown are the calculated band shifts $\chi_{0,\alpha}$ and the renormalized mass m^*_α for $V = 1.55$ eV and $V = 0.46$ eV, respectively. m_e is the free electron mass.

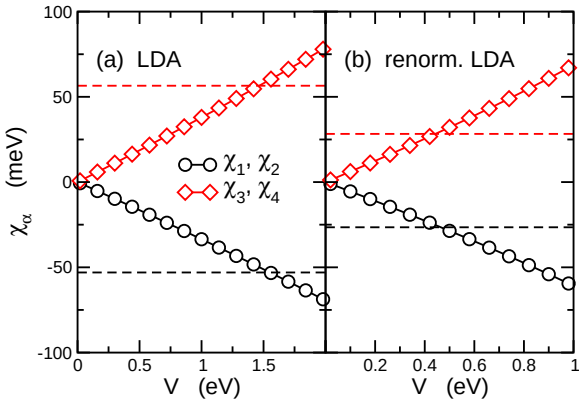

FIG. 2: (Color online). (a) Bands shift $\chi_{0,\alpha}$ as function of the interband coupling V for the parameter set taken from LDA calculations (table I, top). The horizontal dashed lines mark the average band shift as estimated in Ref. [14, 15]; (b) same as in panel (a) but considering LDA band structure renormalized by a factor 2 (table I, bottom).

band edge for each band, namely $E_{\max,1}$, $E_{\max,2}$ for the hole-like bands, $E_{\min,3}$, $E_{\min,4}$ for the electron ones, and we use it to estimate the non-interacting mass for each band $m_\alpha = \hbar^2 k^2_{F,\alpha}/2E_{\max(\min),\alpha}$ and the corresponding density of states $N_\alpha = m_\alpha a^2/2\pi\hbar^2$ (a is here the in-plane lattice constant). Finally, the effective band edge far from the Fermi level [26] follows from the relation $N_\alpha = 1/(E_{\max,\alpha} - E_{\min,\alpha})$. All the estimated values are reported in Table I.

We can now employ a full numerical solution of Eqs. (1)-(2) to get a qualitative estimate the strength of the interband coupling V by comparing the calculated magnitude of the band shifts and the effective mass m^*_α with the experiments. This latter quantity can be evaluated from the numerical solution of Eqs. (1)-(2) as $m^*_\alpha = m_\alpha Z_\alpha$, where $Z_\alpha = 1 - \mathrm{Im}\Sigma_\alpha(i\omega_n)/\omega_n|_{n=0}$. To account for a spin-mediated interaction mechanism we use the Lorentzian spectrum typical of spin fluctuations[27] $B(\Omega) \propto \Omega\omega_0/\pi(\omega_0^2 + \Omega_2)$, with the characteristic energy scale $\omega_0 = 20$ meV[28, 29, 30, 31]. The smallness of ω_0 compared with the Fermi energies, $E_F \approx 100 - 200$ meV (Table I), guarantees the validity of Migdals' theorem [32] also for spin-fluctuations, as discussed e.g. in Refs. [27, 33, 34]. In Fig. 2a we show the band shifts $\chi_{\alpha,0}$ evaluated from the numerical solution of Eqs. (1)-(2) as a function of V by using the set of microscopic parameters obtained by bare LDA calculations. From Fig. 2a we get an estimate $V \approx 1.55$ eV to account for the hole band shift $\Delta_{1,2} \approx 53$ meV reported in Ref. [14]. This value $V \approx 1.55$ eV gives also a theoretical positive shift $\chi_3 = \chi_4 \approx 60$ meV for the electron bands, which agrees quite well with the average experimental value $\bar{\Delta}_{3,4} = (\Delta_3 + \Delta_4)/2 = 56.5$ meV[14]. It is clear that possible differences between the set of hole and electron bands can arise in a more refined treatment when the two electron bands are no more assumed to be degenerate, and when interband coupling is not taken constant. This value $V \approx 1.55$ eV would also give weak-coupling multiband dimensionless interactions $\lambda_{\alpha,\beta} = V_{\alpha,\beta}N_\beta$: $\lambda_{13} = \lambda_{23} = \lambda_{14} = \lambda_{24} = 0.40$, $\lambda_{31} = \lambda_{41} = 0.30$, $\lambda_{32} = \lambda_{42} = 0.58$ which yield effective masses $m^*_\alpha/m_e \approx 1, 1.4, 2$ smaller than what reported in Refs. [14, 15, 16]. A numerical solution of the multiband Eliashberg theory would give also a critical temperature $T_c \approx 29$ K, larger than the experimental one, $T_c \approx 6$ K.

We identify the origin of this discrepancy in the lack, in the above analysis, of high-energy band renormalization pointed out by ARPES experiments[8], which show that the real band structure is roughly twice narrower than the LDA one. The nature of such high-energy renormalization is at the moment unknown, but it must be of different origin than the interband spin-fluctuation coupling which acts on the energy scale ω_0. One can easily see indeed that for a retarded interaction the band renormalization function is negligible $Z(\omega) \sim 1$ when $\omega \gg \omega_0$ [24]. This means that the responsible mechanism for such high-energy renormalization must be operative on an energy scale larger than the electronic bandwidth, suggesting once more a correlation-driven origin [19]. Note also that in this case such mechanism is not expected to affect the band shift since, as shown in Eq. (4), $\chi_{\alpha,0} \approx 0$ for $\omega_0 \gg E_{\max(\min),\alpha}$.

We take into account the bandwidth narrowing by renormalizing the whole LDA band structure as experimentally pointed out by ARPES in LaFePO and $Ba_{0.6}K_{0.4}Fe_2AS_2$. The corresponding microscopic parameters in our model are listed in Table I. Note that, in such rescaled band structure, also the band shifts needed to recover the experimental Fermi areas measured by dHvA techniques are reduced by a corresponding factor 2. The plot of the band shifts as function of the interband coupling, for such renormalized LDA band structure, is shown in Fig. 2b. Our estimate of the interband in-

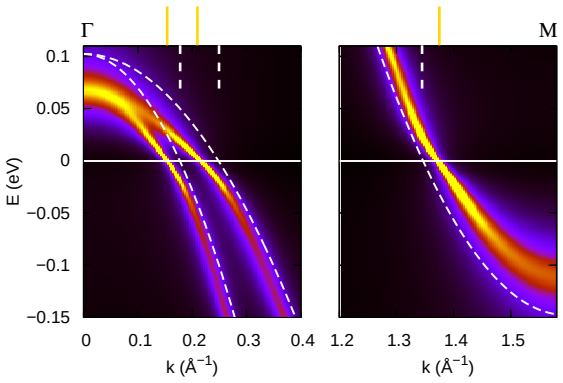

FIG. 3: (Color online). Intensity map of the spectral function for the interacting hole bands (left panel) and the two degenerate electron bands (right panel), obtained by using LDA parameters renormalized by a factor 2 and $V = 0.46$ eV. The dashed lines represent the non-interacting parabolic bands, and the horizontal solid line is the chemical potential. The ticks on the top mark the value of the Fermi vectors in the presence (solid yellow ticks) and in the absence (white dashed ticks) of interaction. Note the shrinking of the Fermi surfaces due to the coupling to the retarded bosonic mode.

teraction in this case is thus $V \approx 0.46$ eV, which gives weak-coupling values $\lambda_{13} = \lambda_{23} = \lambda_{14} = \lambda_{24} = 0.24$, $\lambda_{31} = \lambda_{41} = 0.18$, $\lambda_{32} = \lambda_{42} = 0.34$, with a $T_c \approx 9$ K and $m^*_\alpha/m_e \approx 1.6 - 3.2$, in better agreement with dHvA [14, 15, 16] and specific heat measurements[15, 35, 36]. The corresponding intensity map of the spectral function for the interacting hole and electron bands, as obtained by the Marsiglio-Schossmann-Carbotte analytical continuation [37], is also shown in Fig. 3 along with the bare band dispersions renormalized by the factor 2. Here it is clearly visible the shrinking of the Fermi area of each band due to the interband self-energy effects (χ_α) which give rise to a downward shift of the hole bands and to an upward shift of the electron ones. Note that such self-energy corrections due to the particle-hole asymmetry survive until energies much larger than ω_0, so that the energy shift is effective also at momenta far from the Fermi level. In particular this means that, when interband interactions are predominant, the top of the hole bands (at Γ) and the bottom of the electron bands (at M) approach each other with respect to the prediction of LDA, in agreement with the ARPES observation in 122 compounds[10, 11, 12, 13]. Note also in Fig. 3 the effective mass renormalization at the Fermi velocity $m^*_\alpha = m_\alpha Z_\alpha$ due to the retarded interband interaction. This effect, unlike the band shift χ_α, disappears at the scale energy ω_0, giving rise to a "kink" in the electronic dispersion, recently observed in pnictides [12] and widely discussed in the past in the context of cuprates [38].

In conclusion, we demonstrated that the band shifts reported in pnictides when comparing the experimentally measured Fermi surfaces and band dispersions with LDA calculations are a direct consequence of the coupling to a bosonic mode, once that the strong particle-hole asymmetry and the multiband character of these systems are properly taken into account. Moreover, we showed that the sign of the measured shifts, with both the hole and electron bands approaching the Fermi level, suggests that interband interactions dominate over intraband ones. Our results pose strong constraints on the theoretical modeling of interactions in pnictides, and show unambiguously that finite-band effects cannot be disregarded in these systems.

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